

## Experimental Evidence for a Multicritical Point in the Magnetic Phase Diagram for the Mixed State of Clean, Untwinned $\text{YBa}_2\text{Cu}_3\text{O}_7$

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We report on transport measurements in fields up to 16 T in clean, untwinned single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  which indicate that there is a critical end point for first-order melting in the  $H$ - $T$  phase diagram in the mixed state of that system. Our data show that at a well defined value of the magnetic field, the phase boundary sharply changes slope where the first-order melting transition gives way to a second-order vortex-glass transition. These data suggest that the recently observed first-order transition is robust in the presence of a finite amount of disorder and that the vortex-glass transition only occurs for disorder beyond a certain critical amount.

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The behavior of vortices in the mixed state of the oxide superconductors has been the focus of significant interest and controversy over the last five years [1]. The early suggestions [2-4] of a melting transition as the explanation for the unusual dynamics have been supplemented by more sophisticated ideas [5] about the phase transitions which can occur in these systems in the presence of random pinning. There is now very good experimental [6-11] and theoretical evidence [5,12,13] indicating that in the mixed state in these systems, there is a high-temperature vortex-liquid regime with a finite resistance which is separated from the low-temperature, zero-resistance, ordered phase by a line of phase transitions. In the strong disorder limit, this phase transition is a continuous vortex-glass transition [6,7] while in the weak disorder, clean limit, this transition is a first-order melting transition of the Abrikosov vortex lattice [9]. The mean-field  $H_{c2}$  is a crossover region in the phase diagram separating the normal state at higher temperatures from the vortex-liquid regime at intermediate temperatures.  $H_{c2}$  is not a true phase transition, instead it is where *local* superconductivity and vortices gradually begin to appear upon cooling.

There remain, however, at least two major unresolved issues. The first deals with the importance of the dimensionality of the defects and its influence on the nature of the phase transitions and vortex-glass phase [14]. Disorder in these systems can be zero dimensional as for point defects, one dimensional as in columnar pins (e.g., ion tracks), and two dimensional as is the case for twins. How these various types of defects compete is still largely an open question. The second major unresolved issue is how disorder tunes the transition from the first-order one seen in the clean limit to the continuous transition found in the disordered limit and what the disorder-temperature phase diagram looks like in detail. The issue of how disorder changes the nature of ordered states and the phase transitions into them is a general one in condensed matter

physics. Metal-insulator transitions, Bose glasses, and disordered magnets are other examples of systems which have disorder-tuned phase transitions. The experiments reported here address this second question of how disorder changes the nature of the melting transition in the mixed state of a type II superconductor.

In high-field transport measurements in untwinned, single crystal  $\text{YBa}_2\text{Cu}_3\text{O}_7$  we have found that there is a well defined critical value for the magnetic field above which the first-order melting transition gives way to the continuous vortex-glass transition. At this critical field, the slope of the phase boundary in the  $H$ - $T$  plane changes. Our data then suggest that the first-order transition is robust to a finite amount of disorder and that there is a critical strength for the disorder in the disorder-temperature phase diagram for a type II superconductor, beyond which the transition to the superconducting state is continuous.

Our apparatus is an Oxford Instruments [15] high-field transport cryostat with a variable temperature insert. It allows us to perform measurements in magnetic fields in the range 0-16 T and at temperatures from 1 to 300 K. Access to the high fields that this apparatus allows was crucial to our seeing the effects which we describe below. For the data shown here, we have used impedance-matched normal-state electronics with a phase-sensitive detector operating at 71 Hz. Our measurements typically had a voltage noise of  $6 \times 10^{-9}$  V/ $\sqrt{\text{Hz}}$  and a temperature resolution of 1 mK. Our samples were untwinned single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (YBCO) grown by quenching the tetragonal phase during flux growth [16]. Leads were attached as previously described [9]. These were high-quality, stress-free samples similar to those in which the first-order melting transition has been previously seen. The experiment consisted of measurements of  $\rho_{aa}$  in the low-current, Ohmic regime, as a function of temperature and magnetic field for the field applied parallel to the  $c$  axis of the crystals for two different samples. One sample

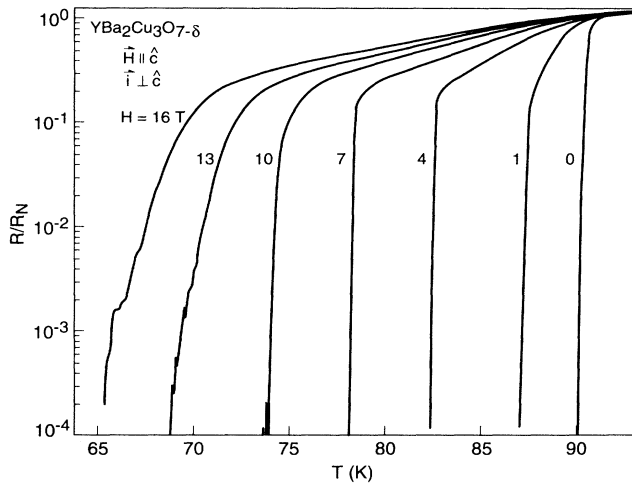


FIG. 1. Normalized linear response resistance as a function of temperature for an untwinned, YBCO single crystal (sample number two) with various values of the magnetic field applied along the  $c$  axis.

has multiple contacts allowing us to measure voltage drops across three different sections of the sample's surface. The two samples and the different sections of sample number two behave quantitatively the same everywhere, except in the details of the jumps in the hysteretic regime.

Shown in Fig. 1 are typical data for this experiment. Plotted is the normalized resistance of sample number two as a function of temperature for fields ranging from 0 to 16 T. The qualitative results are evident from this figure. At low fields, the resistive transitions are sharp and hysteretic with temperature indicating a first-order phase transition. The temperature scale used in this figure is such that one cannot see the hysteresis as the hysteresis widths, when detected, range from 5 to 25 mK. At fields in the range of 8.5 to 11 T, a long low-temperature tail begins to become prominent in the resistance in addition to the sharp drop at higher temperatures. At higher fields the abrupt features have disappeared and one only finds a more gradual behavior of the resistance with temperature. As we will show below, at high fields the transition is continuous and quantitatively agrees with the predictions of the vortex-glass theory. At low fields, the transition is first order and this first-order transition survives up to a finite value of the magnetic field. As we will show below, there is a break in the slope of the phase line where it changes from first order to continuous. This feature marks either a critical end point or a tricritical point in the system.

Shown in Fig. 2 is a plot of the hysteresis widths of our samples as a function of applied magnetic field as measured using two different apparatuses. A typical data set showing hysteresis in the resistive transition is shown as an inset in Fig. 3. The hysteresis widths plotted in Fig. 2 were measured as a function of both temperature and

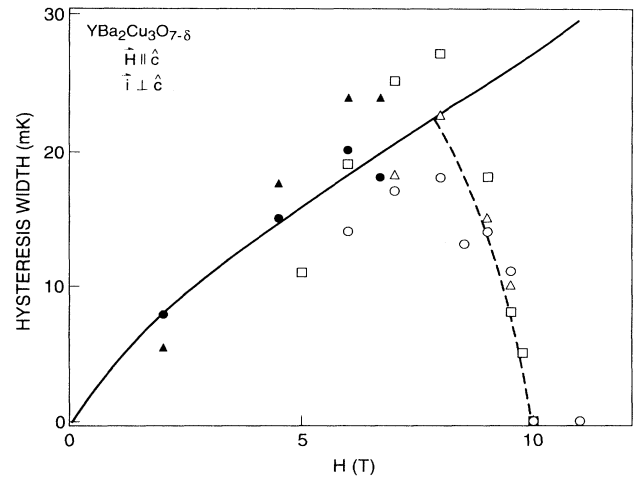


FIG. 2. Widths of the thermal hysteresis loops as a function of applied field as measured with low applied currents in two different systems (full circles, SQUID cryostat, sample 1; open circles, Oxford cryostat, sample 1; open squares, Oxford cryostat, sample 2). Also shown are the independently measured field hysteresis widths converted using the slope of the phase boundary (full triangles, SQUID cryostat, sample 1; open triangles, Oxford cryostat, sample 2). Below 7 T, the hysteresis widths are roughly proportional to the suppression of the melting transition,  $T_m(H)$ , with field, as indicated by the solid line. Note that the hysteresis widths vanish at zero field and at above 10 T.

field on both samples. The widths measured as a function of field were converted to temperature units by using an energy scale set by the local slope of the melting curve [9]. The excellent agreement between thermal and magnetic hysteresis constitutes a very strong evidence for a phase transition. Also shown for comparison is a solid

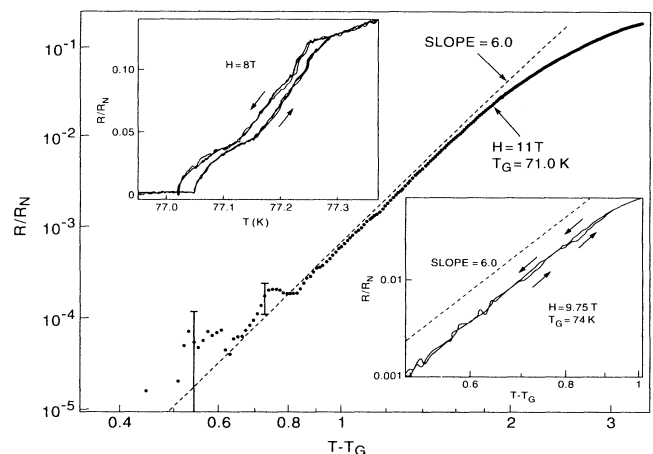


FIG. 3. Log-log plot of the normalized, linear response resistance of sample number two as a function of reduced temperature for a field of 11 T. The data are fitted well by the vortex-glass functional form with an exponent of  $6 \pm 1$ . The insets show the response at 8 and 9.75 T.

line scaled to be proportional to the distance from the zero-field  $T_c$ . For these samples, the hysteresis widths increase with increasing field up to a maximum at roughly 8 T and then decrease again and become zero at 10 T. The data show that the transition is first order for fields less than 10 T and becomes continuous at higher fields. In essence, increasing the magnetic field increases the importance of the disorder in the system. This may occur for at least two reasons: First, the melting transition occurs at lower temperatures for higher fields and the effectiveness of the pinning of vortices to any defect in the system would be expected to increase as the temperature is decreased. Second, increasing the magnetic field also moves the system closer to the quasi-two-dimensional regime, where the interaction between pancake vortices in a given CuO layer are stronger than the interlayer couplings. The two-dimensional system is much more susceptible to disorder: In fact a truly two-dimensional system with random pinning cannot have a first-order phase transition at all [17].

Shown in Fig. 3 is a plot of the resistance of sample number two at three different magnetic fields as a function of temperature. As has been shown previously [6,7], in the disordered limit, the phase transition in this system is well described by the vortex-glass theory. This theory predicts that the linear response resistance will follow the functional form  $\rho \sim (T - T_g)^s$ , where  $T_g$  is the vortex-glass transition temperature and  $s$  is a universal critical exponent. Previous measurements have found  $s \sim 6.5$ . The high-field data of the type shown in Fig. 3 for 11 T follows this functional form quite well with an exponent  $s = 6 \pm 1$ . The transition in the high-field regime is therefore consistent with a continuous, vortex-glass transition.

Shown as insets in Fig. 3 are two data sets at lower fields. The data set for 8 T, plotted on linear scales in the upper left hand corner, shows the well pronounced hysteresis seen for lower fields. Data from three different runs are shown giving an indication of our reproducibility. Note that at low temperatures the resistance transition appears to be sharp, showing no evidence of a resistive tail of the kind found for higher fields.

In the inset to Fig. 3 in the lower right-hand corner is shown a plot of the resistance at an intermediate value of the field (9.75 T). The data show *both* a tail in the resistance at low temperatures which can be fitted by a vortex-glass form and two small hysteretic regions at high temperatures.

Shown in Fig. 4 is a plot of the hysteretic, melting transition temperatures and the vortex-glass transition temperatures (from fits such as shown in Fig. 3) as a function of applied magnetic field for sample number two. In addition are shown contours of constant resistance for the sample. Figure 4 is the central result of this paper. The plots of the transition temperatures show that there is a clear change in the slope and the curvature of the phase boundary where the transition crosses over from being first order to continuous. Also, the equal

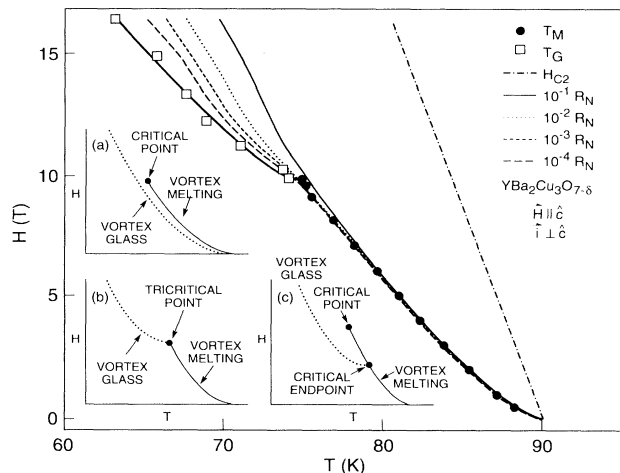


FIG. 4. Composite phase diagram for all of our data on sample number two. The closed circles are the hysteretic melting temperatures  $T_m$ . The open squares are the vortex-glass transition temperatures  $T_g$ . Also shown are  $H_{c2}$  and contours of constant resistance. The insets show three possible phase diagrams which could describe our data. In the insets, the solid curves are lines of first-order transitions, while the dotted curves are continuous transitions. Our data seem most consistent with inset (c).

resistance contours show that the widths of the transitions become much broader at a well defined value of the magnetic field where the change in slope appears to occur. It has been argued [17] that a first-order phase transition should be generally stable to the introduction of weak disorder in three-dimensional systems. The transition is driven continuous only beyond some nonzero disorder strength.

Thus, given that the transition is first order at low fields and that increasing the magnetic field does increase the effective disorder, the magnetic phase diagram in this system should take one of the three forms shown schematically in the insets in Fig. 4. These three possibilities differ in whether and where the line of continuous transitions meets the line of first-order phase transitions. This meeting may (a) never occur, (b) occur at a tricritical point at the end of the first-order line, or (c) occur at a critical end point in the middle of the first-order line. Our data seem most consistent with inset (c) since in a small field range near 9.75 T we see both a hysteretic transition at higher temperatures and a nonhysteretic tail down to lower temperatures. The part of the vortex-liquid regime that intrudes between the two phase transitions as in (a) and (c) has been dubbed "vortex slush" [18].

Shown in the inset to Fig. 3 is a plot of the resistance as a function of temperature for a fixed field of 9.75 T. The resistance curve clearly shows *both* a hysteretic response in the resistance at higher temperatures characteristic of the first-order transition and, at lower tempera-

tures, the long resistive tail which fits the vortex-glass model. Thus there is evidence that for a narrow region around the multicritical point, one can see both transitions as a function of temperature. This suggests that the appropriate phase diagram is the type shown in Fig. 4(c) which has a critical end point and which, for certain values of field over a small range, will show both transitions. The other possibility is that a sample with tricritical point behavior [Fig. 4(b)] and large-scale inhomogeneities could mimic this behavior. The large-scale inhomogeneities in our samples are such that the hysteretic transitions are spread out over a temperature range of  $\sim 0.2$  K as seen in the insets of Fig. 3. Using the slope of the phase boundary, this suggests a tricritical point might be spread in field by roughly 0.2 T, which is also consistent with our data.

In conclusion, we have presented high-field transport measurements in clean, single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_7$ . Our data show that at a well defined value of the magnetic field, the low-field, first-order melting transition gives way to the high-field, continuous vortex-glass behavior. This crossover is accompanied by a change in the slope of the phase boundary. Our experiment suggests that first-order melting is stable in the presence of a small amount of disorder and that there is a critical end point or a tricritical point in the system where the continuous vortex-glass line joins the first-order melting line.

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